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## **Application of Conductive Carbon Nanotube Fibers and Composites: Gas Sensor**

**by Padraig G. Moloney and Enrique V. Barrera**

**ARL-CR-0714**

**May 2013**

**Prepared by**

**Dept. of Mechanical and Materials Science  
Rice University  
PO Box 1892  
Houston TX 77251-1892**

**Under Contract**

**W911NF-11-1-0249**

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# **Army Research Laboratory**

Aberdeen Proving Ground, MD 21005

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14. ABSTRACT Sensing based on carbon nanotube (CNT) materials has been an area of interest since the material's discovery by Iijima in 1991 ( <i>Nature</i> 1991, 354, 56–58), and an active area of research once CNT availability began to increase via the innovations of Smalley and others ( <i>Chem Phys Lett</i> 1995, 243, 49–54). The work presented in this report was conducted at Rice University under contract W911NF-11-1-0249. Here, it is shown that the use of high weight percentage CNT medium density polyethylene (MDPE) composites has suitability towards the sensing of nerve agent stimulants such as dimethyl methylphosphonate (DMMP). The results indicate a possible binary composite approach to sensing, using two variants of CNT. As this work progresses, optimization of the CNT towards sensing DMMP through functionalization is suggested, as is improvement to the experimental apparatus to add functionality, such as controlled and active flow, at a fixed temperature relevant to the application.					
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## **1. Introduction**

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Sensors based on carbon nanotube (CNT) materials have been an area of interest since the material's discovery by Iijima in 1991 (1), and an active area of research once CNT availability began to increase via the innovations of Smalley and others (2, 3). Their long aspect ratios of over 1000-to-1, and varied electrical and thermal properties—along with a decade of research in manipulating nanotubes through functionalization chemistry—make them an ideal target material for sensor research. The scientific community has responded to this opportunity with a wealth of experiments using CNT to sense a wide range of analytes. Some of this work has yielded impressive results in a laboratory setting, such as parts per billion (ppb) sensitivity to chlorine (4), ammonia (5), and nitrogen dioxide (6).

A large body of work in the literature concentrates on hydrogen detection and the detection of water vapor. For relevance to applications of chemical detection, inclusion of these references has been kept to the minimum. Also of note is the large number of research studies that have been conducted in sensing ammonia ( $\text{NH}_3$ ) and nitrous oxides ( $\text{NO}_x$ ). A summary of some of the most promising research in this field is provided in appendix A.

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## **2. Existing Reviews**

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As with all areas of active research, there are a number of pre-existing review articles of relevance to this study. Moore et al. have provided a good review of instrumentation for trace detection of explosives (7). While this does not have direct relevance to nanotechnology, it benefits any research in the sensor area by providing information such as the vapor pressures of various common analytes of interest. Moore makes the useful distinction of breaking down electrochemical sensors in this field into two types, galvanic and electrolytic—a note often ignored by reviews of CNT sensors. A short review by Seto et al. gives a summary of the current sensing technology for nerve and chemical agents (8). This work also includes testing of actual battlefield substances such as sarin, but also sensing of dimethyl methylphosphonate (DMMP) and other common chemicals that closely resemble such agents.

As our research will use composites of CNT and polymers, a useful reference is the Lange et al. review on conductive polymer sensors (9). This work includes a table of electrochemical and electrical results with analyte and polymer. Although of more relevance to biological sensing applications, Rajesh et al. have reviewed conductive polymers, in particular composite applications (10). Included is a summary that highlights analyte, diameter/size, detection limit and voltage among other important data.

With regards to sensor reviews focusing on CNT-based technologies, there are a number of papers of note. Mahar et al. wrote a broad review of CNT sensors, summarizing many of the unique attributes of CNT materials and touching upon sensor applications ranging from temperature to strain (11). They also highlight research that showed changes in Raman spectroscopy data when single wall carbon nanotubes (SWNT) are immersed in various liquids, including common organics (12). In their review, Wang et al. provide a useful summary of specific data for interactions between SWNT and gases (13). These included adsorption energy, charge transfer, and tube-molecule distance.

Bondavalli et al. concentrates on CNT field effect transistors (FET) in their review, a common technical approach taken in CNT sensors (14). They present an informative table on source/drain current change after exposing two types of CNTFET functionalized with DNA sequences to various vapors, including DMMP. Also focused on FET devices is a review by Yáñez-Sedeño (15). This work investigates CNT-metallic nanoparticle electrodes. Rounding out these reviews is one by Jacobs et al. (16), which concentrates on CNT sensors for biomolecules, a broad review by Varghese et al., which includes a useful explanation of sensor nomenclature and basics (17), and Sinha et al., which has some summary of gas sensors (18).

The most useful review work as reference for our research is by Kauffman et al. (19). It comes closest to summarizing the data of interest to our application, including tables that note analyte, method, detection limit, CNT material, and references.

Appendix A summarizes the data from the literature most useful to our research, while reflecting the most useful aspects of the aforementioned CNT sensor reviews.

Research by Boul et al. has previously developed CNT-based sensors for radiation sensing applications for NASA (20). These consisted of networks of SWNT deposited on 32-channel/sensor pads (figures 1 and 2). The SWNT were optimized via functionalization and were designed to register a change in resistivity upon impact from radiation. A change in resistivity will be the primary signal change for our current study.

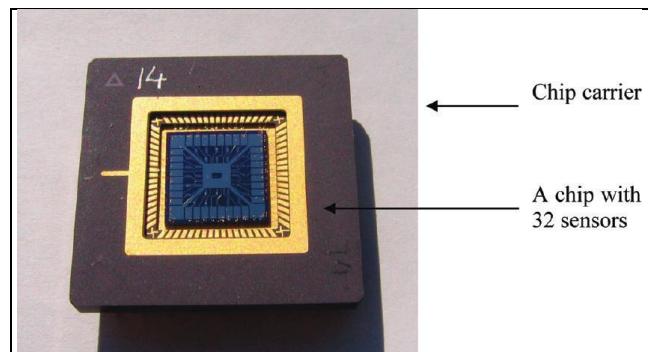


Figure 1. Example of SEM Sensor: 32-channel/sensor pad chip in PLCC 68 chip carrier (20).

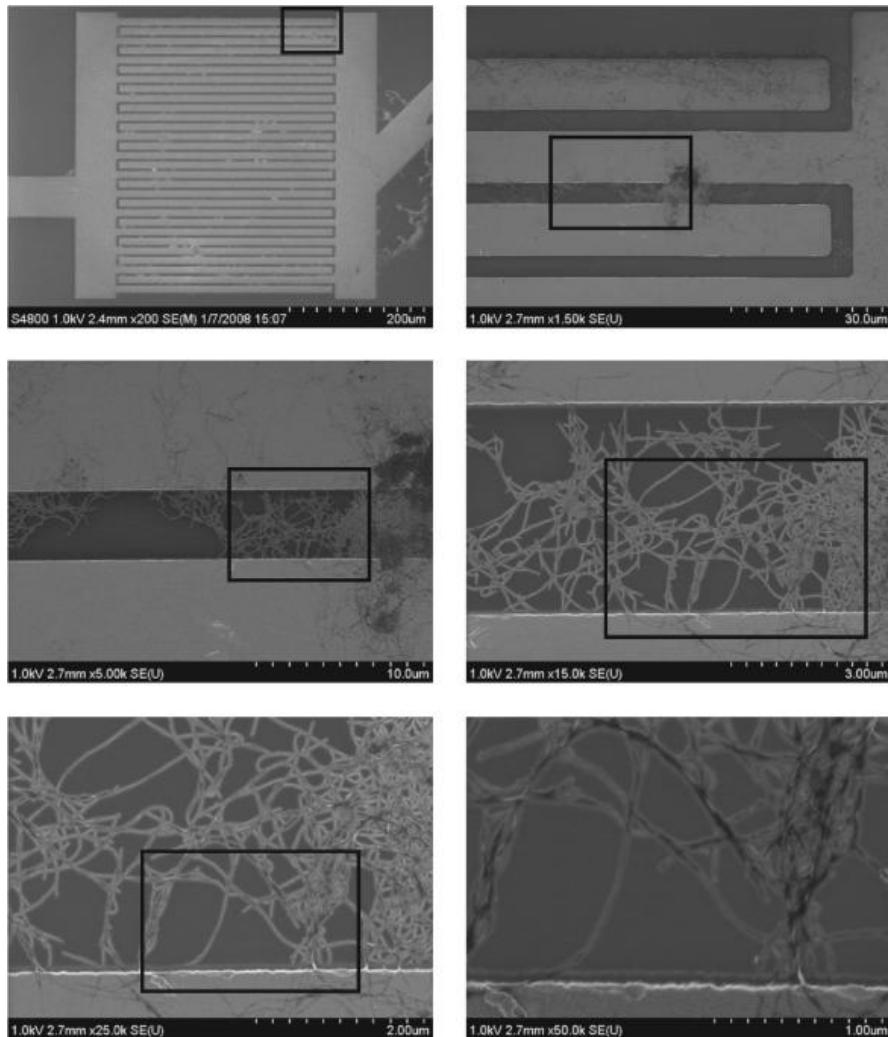


Figure 2. Example of CNT sensor: SEM images (x1000 – x50k) of functionalized SWNT on individual channel/sensor after irradiation with 10 MeV and 30 MeV protons (20).

### 3. Army Applicability – Dimethyl methylphosphonate (DMMP)

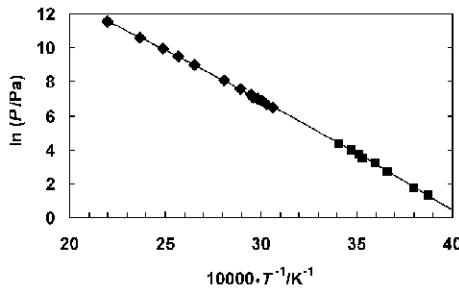
DMMP is of particular interest to our study due to its suitability as a nerve agent stimulant (8, 21). Use of color-changing paper is a commonly used method to test for nerve agents such as sarin (8, 22). The weaknesses of this method are that the agent must be applied directly to the paper in a liquid form, and that the material cannot distinguish between organic solvents, such as DMMP, and actual nerve agents. Instruments such as gas chromatograph-mass spectrometers, surface acoustic wavelength detectors, and ion mobility spectrometers have been produced with a level of portability suitable for limited handheld application (8, 23), but not miniaturized nor automated enough to warrant suitability to low power vehicle applications.

With regards to our initial experiments, a two-fold approach is proposed. Recently produced highly-conductive double wall carbon nanotube (DWNT) fibers will be tested as chemiresistors for DMMP. A follow-on experiment will attempt to modify these fibers with a polymer layer or functionalization to improve selectivity and sensitivity. Secondly, conductive composites of polymer and SWNT will also be tested as chemiresistors of DMMP. An attempt has already been made to modify the porosity and available surface area of these composites to improve their suitability as a gas absorber. DMMP has been obtained from Sigma Aldrich. A colorless liquid, it has a boiling point of 181 °C, a vapor pressure of 160 Pa at 25 °C, vapor density of 4.2, and a liquid density of 1.15 g/mL (24, 25). The flammable flash point is 69 °C. Calculated and measured vapor pressure data in a temperature range from 263 °K (-10 °C) to 453.8 °K (180.65 °C) can be found in the literature (25, 26) (figure 3).

**Table 3. Measured Vapor Pressures for DMMP, Calculated Values Based on Antoine Coefficients listed in Table 9, and Percent Difference**

T K	P Pa	P <sub>calc</sub>	difference <sup>a</sup> %
		Pa	%
258.2	3.73	3.67	1.8
263.4	5.67	6.20	- 8.6
273.2	15.2	15.2	- 0.1
278.2	24.5	23.5	4.4
283.2	35.0	35.5	- 1.4
285.0	41.7	41.0	1.7
288.2	52.8	52.8	0.1
293.2	78.5	77.2	1.8
326.4	653.3	677.3	- 3.8
330.2	799.9	840.3	- 4.8
332.4	933.3	949.5	- 1.7
335.2	1067	1112	- 4.1
337.6	1200	1265	- 5.1
339.2	1387	1376	1.0
345.8	1973	1921	2.7
356.8	3346	3271	2.3
376.8	7999	7880	1.5
389.8	13 350	13 160	1.4
402.0	21 280	20 580	3.4
421.8	39 930	39 920	0.0
454.4	102 200	102 800	- 0.6

<sup>a</sup> Percent difference was calculated as  $100(P_{\text{meas}} - P_{\text{calc}})/P_{\text{calc}}$ .



**Figure 3.** DMMP vapor pressure: [ , data generated by DSC methodology; 9 , data generated by saturator methodology; s , Antoine correlation equation.

**Table 4. Calculated Vapor Pressure, Volatility, and Enthalpy of Vaporization for DMMP Based on Antoine Coefficients Listed in Table 9**

T K	P Pa	volatility	$\Delta_{\text{vap}}H$
		mg · m⁻³	kJ · mol⁻¹
263.2	6.025	341.8	55.9
273.2	15.22	831.8	54.9
283.2	35.49	1871	54.0
293.2	77.15	3928	53.2
298.2	111.1	5562	52.8
303.2	157.7	7763	52.4
313.2	305.1	14 540	51.8
333.2	992.2	44 450	50.6
353.2	2760	116 600	49.5
373.2	6758	270 300	48.6
393.2	14 900	565 700	47.8
413.2	30 100	1 088 000	47.1
433.2	56 490	1 947 000	46.5
453.8	101 325	3 333 000	46.0

Figure 3. Measured and calculated vapor pressures for DMMP (25).

An optimized nanotube sensor could offer a solution for sensing a nerve agent simulant such as DMMP. Picking some of the most relevant research from the literature and focusing on the chemiresistive sensor approach, figure 4 illustrates chemical sensitivity to DMMP versus

resistance change. One can note that an enormous resistance change is not produced in any of this previous research; these small-realized signal changes may make applicability difficult.

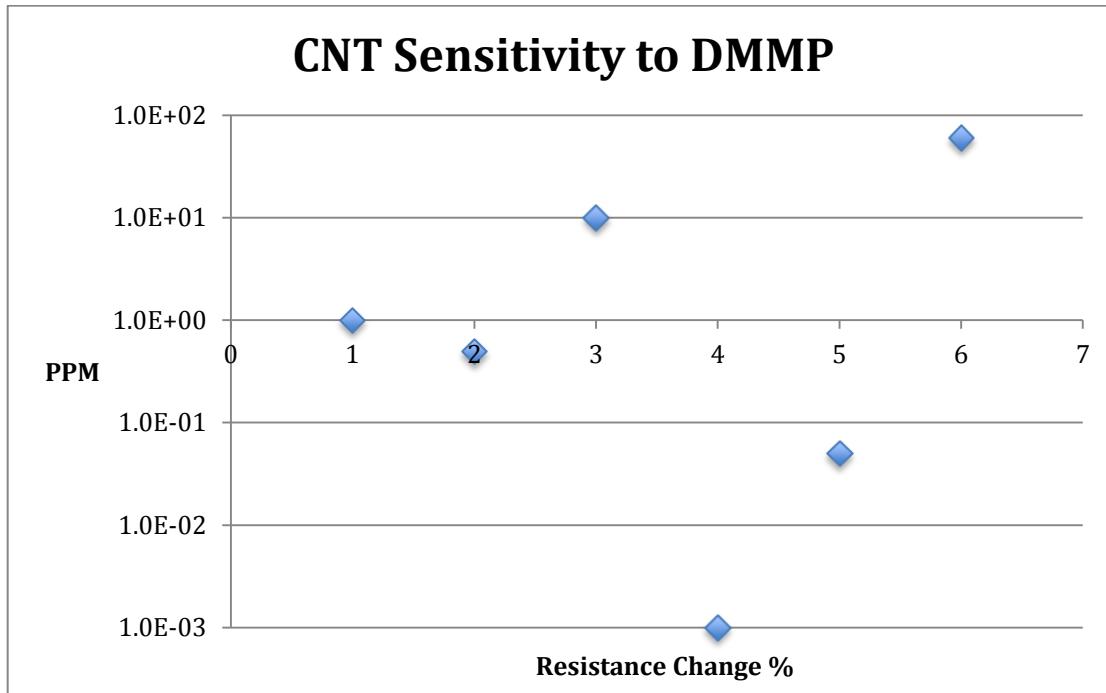


Figure 4. Summary of select research: sensitivity vs. relative resistance change of CNT-based DMMP sensors.

A variety of voltage levels were also found throughout the literature to “drive” such CNT sensors of DMMP. These range from 50 mV (27), 0.1 V (28) to 3 V (29). Such voltage levels should be taken into consideration when reflecting on the proposed application of such a sensor towards low-power applications. It is hoped that the use of conductive nanotube fibers and composites in this study will reduce the overall power needs when compared to previous studies, while increasing the fidelity of signal change.

Observing relevant research in the literature, one can note a number of experimental approaches. The experimental setup as illustrated by Choi et al. represents what is probably the most optimal (30) (see figure 5) and should be given consideration for future work. However, given the limited amount of time and resources available for our study, a less complex test setup will be used. Instead, we will use an approach similar to a simple one found in the literature (31)—a glass vessel with a Teflon-taped plastic lid, combined with the appropriate outlets for vacuum, wiring, and syringe (to introduce the DMMP). Given the restrictions of the multimeter, only one sample/sensor will be tested at a given time.

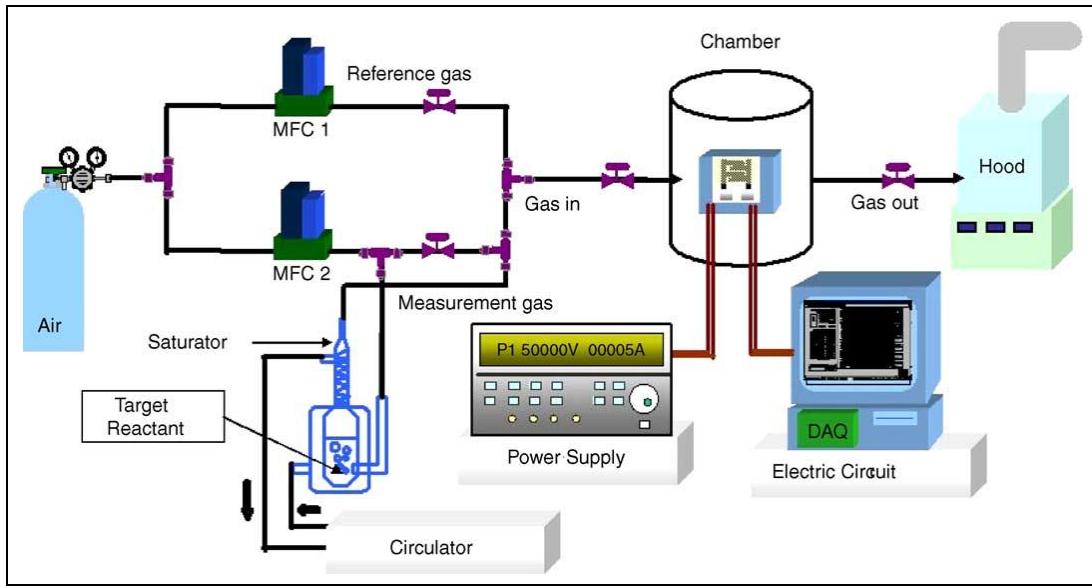


Figure 5. One possible experimental setup (30).

Novak et al. used a permeation tube and mass flow controllers to deliver calibrated doses of DMMP (29). They also improved the sensitivity of their sensor by applying a 100 nm coating of an acidic strong hydrogen-bonding polycarbosilane, which acted as a chemical-selective polymer. In follow-on research, Snow and Novak improved the sensitivity further by changing to a capacitive rather than resistive response and achieved sensitivity in the ppb level (32). They also observed that DMMP absorbs onto SWNT, and is a strong electron donor, causing a transfer of negative charge, which in turn changes transistor threshold voltage by  $-2\text{ V}$  (29). This work also showed that after an initial response of 10+ min, it took many hours for the sensor to fully recover as DMMP desorbs slowly from SWNT. An additional positive charge can be applied to offset the negative resulting from DMMP adsorption and speed up recovery of the device.

Wang et al. also took a composite-style approach, using hexafluoroisopropanol-substituted polythiophene (HFIP-PT) and poly(3-hexylthiophene) (P3HT)—SWNT spin-coated films as chemiresistors (28). They chose the HFIP group to polythiophene because of its H-binding with phosphate esters. These esters are common in chemical warfare agents, including sarin gas and stimulants such as DMMP. They showed sensitivity as low as 0.6 ppm and responses as fast as 1–2 min.

Kyle produced network films of SWNT on substrates of polyethylene terephthalate (PET) (21). This work is highly relevant from a composite standpoint to ours. They filter out interfering vapors such as hexane by using a 2-micron thick barrier film of chemical-selective polymer polyisobutylene (PIB) on the SWNT surface. Their experiment was at room temperature. A major advantage of their work is a large resistance swing of 75–100%, albeit at a response time measured in the 10's of minutes. Also, one should note that their experiments were conducted solely in the presence of DMMP, with no other gas present.

Our approach of testing both our conductive DWNT fibers and conductive SWNT-polymer composites offers a potentially successful path to a DMMP sensor. Modification of both with either a polymer layer and/or functionalization also has the promise to improve selectivity and sensitivity.

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#### **4. Initial Experiments – Composite DMMP Sensor**

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Early stage experiments were carried out using a sealed 2-L glass vessel, with air at ambient conditions. Small droplets of DMMP were placed at the bottom of the vessel to simulate various ppm conditions. The composites were placed on a four-point probe (Cascade C4S), sealed in the glass vessel, and instrumented with a multimeter (Agilent 34410A). The vessel was then placed on a hot plate to evaporate the analyte. Temperatures near the sample and the hot plate were monitored using thermocouples.

High weight percentage composites of both SWNT (HiPco, Rice University) and MWNT (Mitsui) in medium density polyethylene (MDPE) were tested. The SWNT nanotubes were purified via a simple 1-pot method (33). The MWNT were received in a highly pure state. The composites were prepared via a modified version of a simple solvent blend technique (34). These modifications will be discussed in a future publication. Percent loadings of 90% SWNT were used for these tests, as they produce the lowest electrical resistivity, better than  $1E-03 \Omega\cdot cm$ .

Data from these initial experiments hold great promise. Figure 6 shows the results of 1000 ppm DMMP exposure on the MWNT composites. Upon introduction of DMMP vapor into the air inside the vessel, the composites show an increase in resistivity of almost 30% within minutes, and a recovery response within 5–15 min. It should be noted that the atmosphere inside the vessel is static; an optimal testing regime would include active mixing of the analyte and controlled flow, as seen in figure 5. With that in consideration, these results here should be seen as conservative.

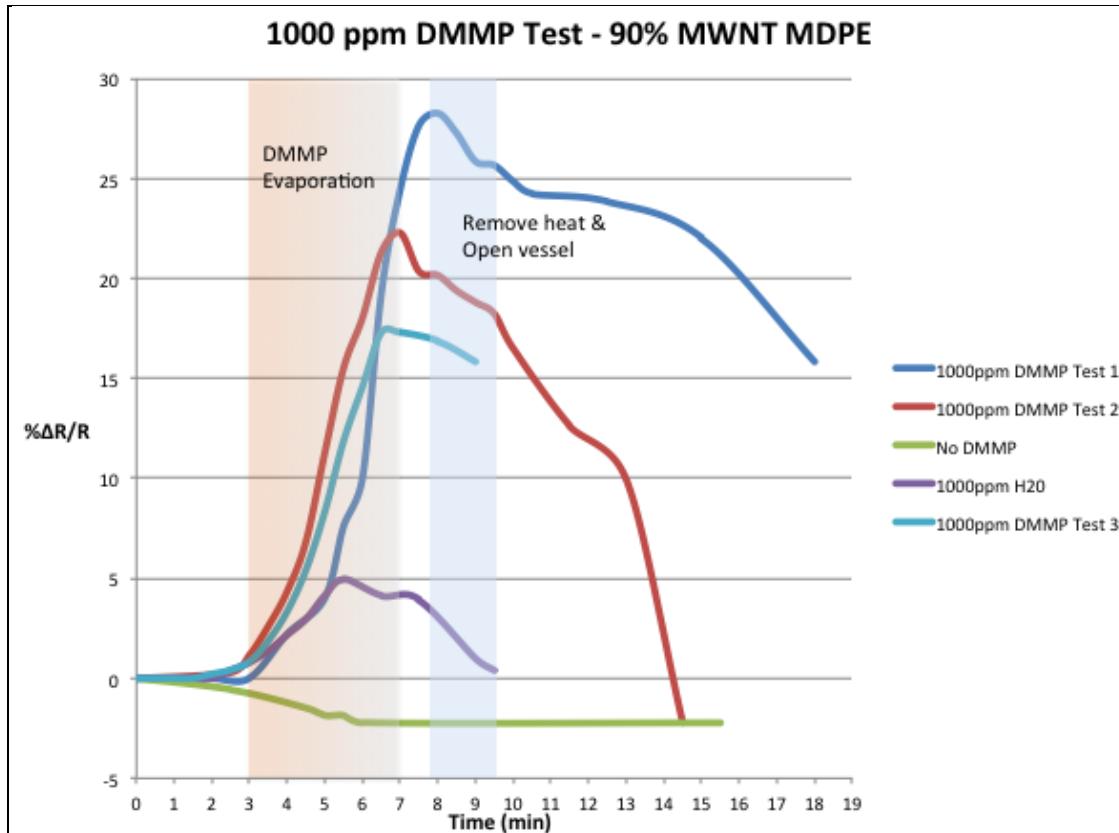


Figure 6. DMMP sensing tests with MWNT MDPE composites.

The first of two possible sources of error were then considered for this experimental setup—radiative heating. Tests were performed that showed negligible effect from radiative heating on either the MWNT or SWNT samples, shown as “no dmmp” for both data sets. A resistance change of 2% can be attributed to radiative heating for the MWNT samples and 1% for the SWNT samples.

The SWNT tests also showed interesting performance under the exposure to DMMP. One thousand ppm testing showed response times in minutes and a resistance change of up to 15% (figure 7). Repeatability under multiple cycles was not as impressive with the SWNT samples versus the MWNT. However, it is notable that the resistance of this composite behaves in a different manner to the MWNT equivalent. Testing showed that it decreased in resistivity upon exposure to DMMP, rather than an increase, as with MWNT composite. This distinction holds great promise, as both composites could be used as a reliable binary confirmation of the presence of DMMP. We are still researching why the two composites behave so differently. The mechanism of DMMP molecule binding to CNT is still not well understood. DMMP is a strong electron donor and this will be considered as research progresses.

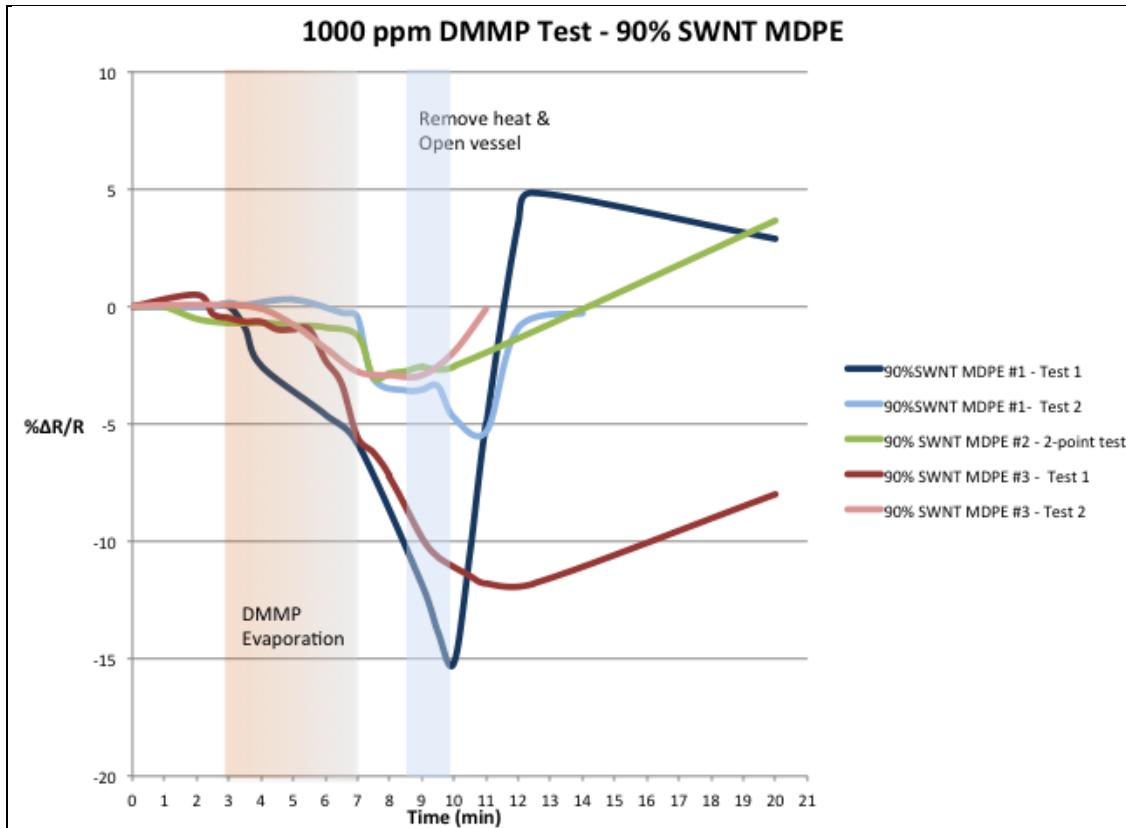


Figure 7. DMMP sensing tests with SWNT MDPE composites.

The SWNT composite showed a higher sensitivity to both convective heating and water vapor than the MWNT. Figure 8 shows this sensitivity. It should be noted that while opening the vessel and exposing the composites to fresh air contributed to signal recovery, the thermocouples showed that the heating effect on the samples and four-point probe was slower to abate. This leads us to believe that it is the presence of vapor that is causing the initial change in resistance, rather than any thermal effect. Further testing with additional solvents will be conducted to assess the proportion of signal change that can be attributed to either the water vapor or the convective heating.

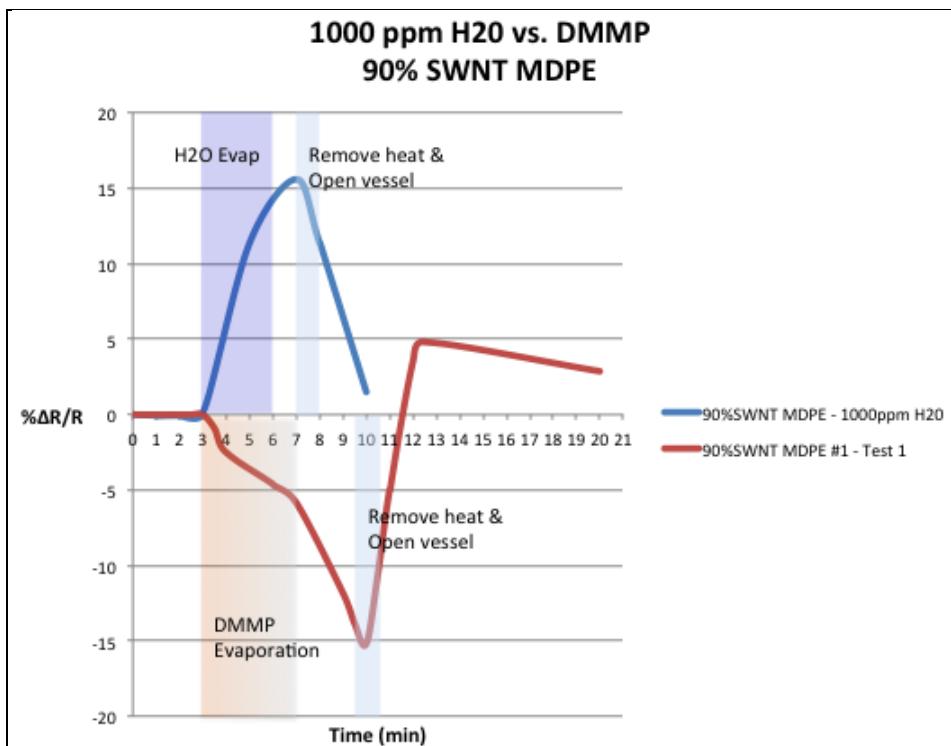


Figure 8. Water vs. DMMP.

Surface area and porosity analysis through BET was performed on the SWNT composites. Composites exposed to boiling dichlorobenzene, to increase porosity and surface area, showed no large surface area change. The surface area of these composites remains at  $100\text{--}200 \text{ m}^2\text{g}^{-1}$  and showed no marked difference in DMMP sensitivity. Research on the pore size optimization of the composite is ongoing.

Additional experiments were performed on the SWNT composites to explore the lower bounds of DMMP sensitivity. Given the aforementioned simplicity and weaknesses of this initial experimental protocol, these data should be considered conservative. The 10 and 100 ppm tests in figure 9 show a positive change in resistivity. At these lower ppm, one can see that radiative and convective heating effects associated with this test, while ineffectual at 1000 ppm, are now dominant and mask the sensing of the DMMP, itself. Only at 500 ppm does the DMMP sensing become dominant. These effects will have to be considered during future research and design of a more mature test apparatus.

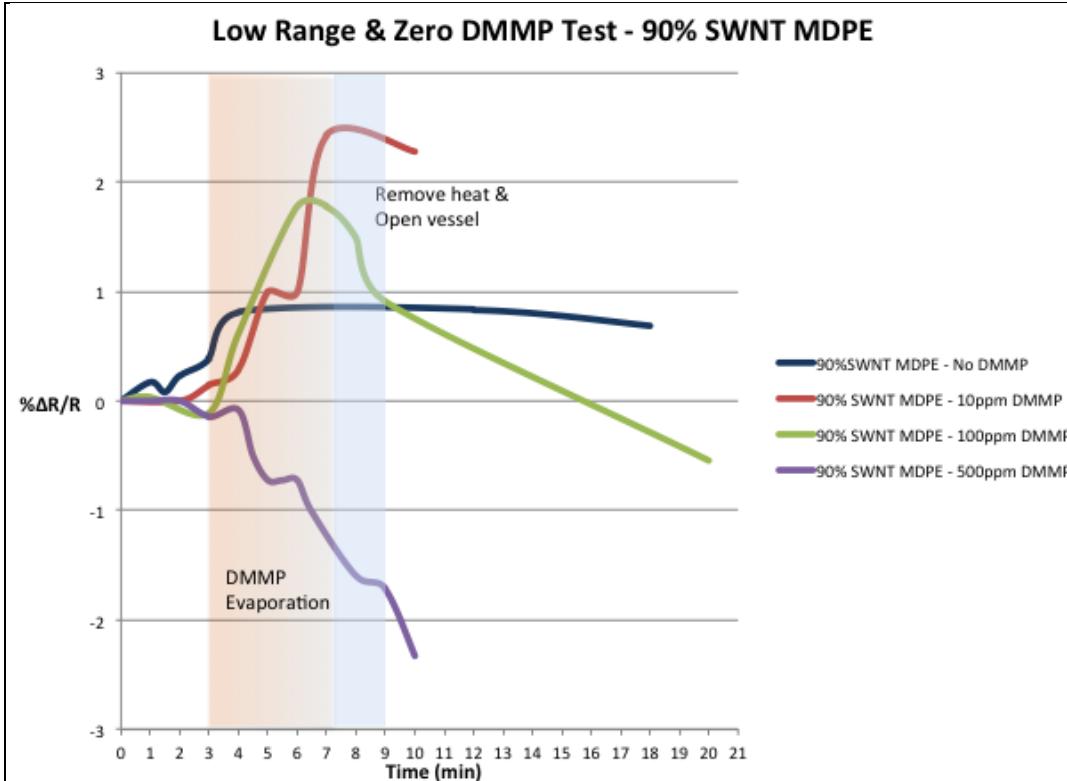


Figure 9. Low ppm exposure tests SWNT MDPE composite to DMMP.

In conclusion, the use of high weight percentage CNT MDPE composites has suitability towards the sensing of nerve agent simulants such as DMMP. This early stage work has created a possible binary composite approach to sensing, using two variants of CNT. Further testing of these composites will continue, in addition to testing of the solid CNT fibers. As this work progresses, optimization of the CNT towards sensing DMMP through functionalization is suggested, as is improvement to the experimental apparatus to add functionality, such as controlled and active flow, at a fixed temperature relevant to the application.

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## Appendix A.

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Summary of the experimental elements of reviewed publications, arranged alphabetically with respect to the analyte being targeted by the sensor.

Year	Author	Analyte	CNT Type	Functionalized/Matrix	Enviro	Method	Detection Limit	Ref.
2006	Robinson	Acetone, MeOH	SWNT	Oxidation	Gas	Resistance, Capacitance	--	(35)
2005	Anand	Air	SWNT	--	Gas	Resonant Cavity	--	(36)
2004	Chung	Air, oxygen	MWNT	--	Gas	Voltage	--	(37)
2003	Someya	Alcohols	SWNT	--	Gas	Current	--	(38)
2006	Kim	Ar	MWNT	--	Gas	Voltage	--	(39)
2010	Leghrib	Benzene	MWNT	Rh, Pt, Au, Pd, Ni	Gas	Resistance	50 ppb	(40)
2004	Cho	Benzene, ethanol	SWNT	Ethyl cellulose	Gas	Resistance	1000 ppm	(41)
2009	Mabrook	C2H5OH	SWNT	Polyelectrolyte inkjet	Gas	Current	300 ppm	(42)
2002	Yang	CH3OH, C2H5OH	SWNT	-- poly(3-methylthiophene)	Gas	Absorbance	--	(43)
2005	Santhanam	Chloromethane	MWNT		Gas	Resistance	5 ppm	(44)
2011	Gohier	Cl2	MWNT	PEI	Gas	Resistance	27 ppb	(4)
2004	He	CO	MWNT	HClO4	Gas	Cyclic volt	--	(45)
2006	Wanna	CO	MWNT	PANI	Gas	Resistance	150 ppm	(46)
2007	Santhosh	CO	MWNT	PDPA	Gas, Liquid	Voltage	0.01 ppm	(47)
2008	Wongwiriyapan	CO	SWNT	Pt	Gas	Resistance	1 ppm	(48)
2010	Kauffman	CO	SWNT	Au	Gas	Modeling Paper		(49)
2007	Kuzmych	CO, CO2, O2	SWNT	PEI	Gas	Resistance	5 ppb	(50)
2001	Ong	CO2	MWNT	SiO2	Gas	Permittivity	--	(51)
2004	Star	CO2	SWNT	PEI, Starch	Gas	Current	--	(52)
2005	Zribi	CO2	SWNT	--	Gas	Vibrational Resonance	--	(53)
2003	Philip	Dichloromethane, acetone, chloroform	SWNT	Oxidation, PMMA	Gas	Resistance	--	(54)
2004	Jose	Dichloromethane, acetone, chloroform	MWNT	PMMA	Gas	Resistance	--	(55)
2010	Wang	DMMP	SWNT	PEI	Gas	Resistance	1 ppm	(56)
2011	Wang	DMMP	SWNT	CoPc	Gas	Resistance	0.5 ppm	(27)
2001	Hopkins	DMMP, DIMP	--	Carbon Black/Polymer	Gas	Resistance	--	(57)
2006	Kyle	DMMP, DIMP	SWNT	PET, PIB	Gas	Resistance	299 ppm	(21)
2005	Choi	DMMP, DPGME, DCM	--	Al2O3, In2O3, Pt, Pd, ZnO, ZrO2	Gas	Resistance	0.5 ppm	(30)
2003	Novak	DMMP, Hexanes, Xylenes, H2O	SWNT	--	Gas	Resistance	--	(29)
2008	Wang	DMMP, VOC	SWNT	HFIP-PT, P3HT	Gas	Voltage, Resistance	0.05 ppm	(28)
2007	Ali	Dopamine	SWNT	PABA	Liquid	Cyclic volt, diff pulse volt	40 pM, 1 nM	(58)

2011	Delalande	DPCP	SWNT	Au	Gas	Voltage	1 ppm	(59)
2006	Yujin	Ethanol	MWNT	SnO2	Gas	Resistance	50 ppm	(60)
2008	Krishna	H2	SWNT	Pd	Gas	Resistance	--	(61)
2011	Zilli	H2	MWNT	Pd	Gas	Resistance	70 ppm	(62)
1999	Liu	H2	SWNT					(63)
2008	Gong	H2	SWNT	SnO2	Gas	Resistance	--	(64)
2001	Kong	H2	SWNT	Pd	Gas	Resistance	40 ppm	(65)
2005	Sayago	H2	SWNT	Pd	Gas	Resistance	--	(66)
2007	Sayago	H2	SWNT	Pd	Gas	Resistance	--	(67)
2009	Srivastava	H2	MWNT	PANI	Gas	Resistance	--	(68)
2003	Wong	H2	MWNT	Pd	Gas	Voltage	--	(69)
2005	Sippel-Oakley	H2	SWNT	Pd	Gas	Resistance	10 ppm	(70)
			MWNT					
2004	Ahn	H2, NH3	Forest	--	Gas	D.C.	8 ppm	(71)
2006	Star	H2, CH4, CO, H2S	SWNT	Pd, Pt, Rh, Au	Gas	Voltage	5 ppm	(72)
		H20, acid, MeOH						
2005	Staii	DMMP etc.	SWNT	DNA	Gas	Current	25 ppm	(73)
2010	Bekyarova	H2Cl	SWNT					(74)
2000	Zahab	H2O	SWNT		Gas	Resistance	--	(75)
2003	Kim	H2O	SWNT	PMMA	Gas	Voltage	--	(76)
2004	Star	H2O	SWNT	Nafion	Gas	Current	--	(77)
					Quartz			
2005	Chen	H2O	SWNT	Nafion	Gas	microbalance	15.76	(78)
2005	Na	H2O	SWNT	--	Gas	Voltage	--	(79)
2006	Su	H2O	SWNT	PAMPS-SiO2	Gas	Impedance	--	(80)
2006	Yu	H2O	MWNT	PEI	Gas	Resistance	--	(81)
2007	Jiang	H2O	MWNT		Liquid	Resistance	--	(82)
2007	Su	H2O	MWNT	PMMA, KOH	Gas	Impedance	--	(83)
2011	Tang	H2O	MWNT	PI	Gas	Resistance	--	(84)
					Voltage,			
2003	Wang	H2O2, NADH	SWNT	Teflon	Liquid	Resistance	--	(85)
2011	Izadi	H2S	MWNT	Carboxyl, amide, Mo, Pt	Gas	Resistance	--	(86)
2009	Fam	H2S, CO, NO	SWNT	Ag	Gas	Current	2 ppm	(87)
2004	Valentini	HCl	MWNT	POAS	Gas	Resistance	100 ppm	(88)
2006	Li	HCl, Cl2	SWNT	CSPE, HPC	Gas	Resistance	2 ppm	(89)
2006	Zhang	HCN	SWNT	Boron modeling paper	Gas	--	--	(90)
2006	Wang	HCOH	SWNT	Modeling paper	--	--	--	(90)
2010	Khani	Hg(II)	MWNT	BMIM.BF4	Liquid	Voltage	0.5 ppb	(91)
2010	Lee	Hg2+	SWNT	--	Liquid	Resistance	--	(92)
2006	Liu	LPG, C2H5OH	MWNT	SnO2	Gas	Voltage	10 ppm	(93)
1989	Bartlett	MeOH	--	Gold Mylar Layers	Gas	Resistance	--	(94)
2010	Chen	MeOH, IPA	SWNT	DNA	Gas	Resistance	--	(95)
2004	Valentini	Methane	MWNT		Gas	Resistance	--	(96)
2004	Arab	Modeled Various	SWNT	--	Gas	--	--	(97)

2010	Tooski.	Modeled Various	--	--	--	--	--	(98)	
2003	Bradley	--	CNT	NaPSS polymer	Gas	Voltage	--	(99)	
2009	Loh	--	SWNT	Ni	Gas	Voltage	--	(100)	
2005	Huang	N2	MWNT	--	Gas	Resistance	--	(101)	
2000	Sumanasekera	N2, He	SWNT	--	Gas	Resistance	--	(102)	
2001	Varghese	NH3	MWNT	SiO2	Gas	Impedance	--	(103)	
2002	Chopra	NH3	SWNT	Epoxy conductive	Gas	Microwave Resonance	100 ppm	(104)	
2003	Bradley	NH3	SWNT		Liquid/Gas	Voltage	--	(105)	
2003	Bradley	NH3	SWNT	PEI	Gas	Voltage	--	(106)	
2003	Modi	NH3	MWNT	--	Gas	Voltage	--	(107)	
2003	Suehiro	NH3	MWNT		Gas	Resistance	10 ppm	(108)	
2004	Bekyarova	NH3	SWNT	PABS	Gas	Resistance	5 ppm	(109)	
2004	Jang	NH3	MWNT		Gas	Resistance	--	(110)	
2004	Lu	NH3	SWNT	Pd	Gas	Resistance	6 ppm	(111)	
2004	Wang	NH3	MWNT	--	Gas	Resistance	>10 ppm	(112)	
2005	Feng	NH3	SWNT	HNO3	Gas	Absorbance	--	(113)	
2006	Li	NH3	MWNT	--	Gas	Resistance	50 ppm	(114)	
2006	Nguyen	NH3	SWNT	MWNT	--	Gas	Resistance	5 ppm	(115)
2006	Quang	NH3	SWNT		Gas	Resistance	5 ppm	(116)	
2006	Zhang	NH3	SWNT	PANI	Gas	Resistance	50 ppb	(117)	
2007	Duc Hoa	NH3	SWNT	SnO2	Gas	Resistance	10 ppm	(118)	
2008	Van Hieu	NH3	MWNT	SnO2	Gas	Resistance	>100 ppm	(119)	
2009	He	NH3	MWNT	PANI	Gas	Resistance	<12 ppm	(120)	
2009	Zhang	NH3	SWNT	PANI(CSA)	Gas	Resistance	10 ppb	(5)	
2010	Lim	NH3	SWNT	--	Gas	Resistance	1 ppm	(121)	
2010	Mangu	NH3	MWNT	alumina	Gas	Resistance	--	(122)	
2009	Peng .	NH3	SWNT	SiO2	Gas	Resistance	<100 ppm	(123)	
2004	Villalpando-Perez	NH3, acetone, OH	--	Modeling paper	--	--	--	(124)	
2003	Chopra	NH3, CO, Ar, N2, O2	SWNT	Epoxy conductive	Gas	Dielectric	100 ppm	(125)	
2006	Bittencourt	NH3, CO, NO2	MWNT	WO3	Gas	Resistance	100 ppm, 10 ppm, 500 ppb	(126)	
2006	Jones	NH3, EtOH, H2O	SWNT	DNA, ACDEP	Gas	Voltage	--	(127)	
2007	Li	NH3, MeOH	MWNT	(NH2OH)(HCL), PMMA	Gas	Voltage	--	(128)	
2006	Ma	NH3, N(CH3)3, Et3N	MWNT	PANI	Gas	Current	--	(129)	
2007	Zhang	NH3, NO2	SWNT	PABS	Gas	Resistance	100 ppb, 20 ppb	(130)	
2011	Mangu	NH3, NO2	MWNT	PEDOT:PSS, PANI	Gas	Resistance	100 ppm	(131)	
2010	Lim .	NH3, NO2, H2S	SWNT	PANI	Gas	Resistance	50, 500, and 500 ppb	(121)	

2007	Terranova	NH3, NOx	SWNT	--	Gas	Resistance	75 ppm	(132)
2002	Ong	NH3, O2, CO2	MWNT	SiO2	Gas	Permittivity	--	(133)
2007	Tabib-Azar	NH3OH, HCl	MWNT	--	Gas	voltage	--	(134)
2007	Maklin	NO	SWNT	carboxyl	Gas	Resistance	100 ppm	(135)
2011	Li	NO CO	SWNT	Pd, PT	Gas	Modeling Paper		(136)
2009	Hoa	NO, NH3	SWNT	--	Gas	Resistance	2 ppm	(137)
2003	Cantalini	NO2	MWNT	--	Gas	Resistance	10 ppb	(138)
2003	Valentini	NO2	MWNT	--	Gas	Resistance	<10 ppb	(139)
2004	An	NO2	SWNT	Ppy	Gas	Resistance	5 ppm (extrapolated)	(140)
2004	Cantalini	NO2	MWNT	--	Gas	Resistance	10 ppb	(141)
2004	Valentini	NO2	MWNT	--	Gas	Resistance	10 ppb	(142)
2005	Mercuri	NO2	SWNT	--	Gas	Photoemission	--	(143)
2005	Young	NO2	SWNT	MPCs	Gas	Current	4.6 ppb	(6)
2006	Larciprete	NO2	SWNT	Rh	Gas	Photoemission	--	(144)
2006	Suehiro	NO2	MWNT	Al , Cr	Gas	Resistance	0.5 ppm	(145)
2006	Wongwiriyapan	NO2	SWNT	--	Gas	Resistance	200 ppb	(146)
2006	Zhang	NO2	SWNT	PMMA, SU-8	Gas	Resistance	20 ppm	(147)
2007	Larciprete	NO2	SWNT	Rh	Gas	Photoemission	--	(148)
2008	Bal·zsi	NO2	MWNT	hex-WO3, Ag Au	Gas	Resistance	100 ppb	(149)
2008	Lee	NO2	SWNT	--	Gas	Resistance	3 ppm	(150)
2008	Moon	NO2	MWNT	Binder	Gas	Voltage	50 ppm	(151)
2011	Leghrib	NO2	MWNT	B, N, SnO2,	Gas	Resistance	100 ppb	(152)
2004	Wei	NO2	SWNT	SnO2	Gas	Resistance	200 ppm	(153)
2004	Liang	NO2,	MWNT	SnO2	Gas	Resistance	2 ppm	(154)
2003	Santucci	NO2, CO	MWNT	--	Gas	Resistance	10 ppb	(155)
2003	Cantalini	NO2, CO, NH3, EtOH, C6H6, H20	MWNT	--	Gas	Resistance	10 ppb (NO2)	(138)
2004	Valentini	NO2, CO, NH3, H2O, C2H5OH	MWNT	--	Gas	Resistance	10 ppb	(142)
2006	Lu	NO2, HCN, HCl, Cl-2, acetone, benzene	SWNT	Metal, polymer coatings	Gas	Resistance	Various ppm	(156)
2000	Kong	NO2, NH3	SWNT	Semiconducting Tubes	Gas	Resistance	200 ppm	(157)
2003	Qi	NO2, NH3	SWNT	Nafion, PEI	Gas	Resistance	<1 ppb	(158)
2005	Li	NO2, NH3	SWNT	--	Gas	Resistance		(159)
2007	Jung	NO2, NH3	MWNT	--	Gas	Resistance	100 ppb	(160)
2007	Penza	NO2, NH3	MWNT	--	Gas	Resistance	10 ppm	(161)
2007	Penza	NO2, NH3	MWNT	Au, Pt	Gas	Resistance	100 ppb	(162)
2008	Penza	NO2, NH3, H2S	MWNT	Pt, Pd	Gas	Resistance	<1 ppm	(163)
2003	Li	NO2, nitrotoluene	SWNT	--	Gas	Resistance	<44 ppb	(164)
2003	Goldoni	NO2, SO2, NH3	SWNT	--	Gas	Photoemission	<10 ppb	(165)
2004	Goldoni	NO2, SO2, NO	SWNT	--	Gas	Photoemission	<10 ppb	(166)

2008	Ueda	NOx	MWNT	--	Gas	Resistance	2 ppm	(167)
2008	Ueda	NOx	MWNT	laser	Gas	Resistance	--	(168)
2008	Ueda	NOx	SWNT, MWNT	--	Gas	Resistance	5 ppm	(169)
2000	Collins	O2	SWNT	--	Gas	Voltage	--	(170)
2001	Wadhawan	O2, Ar, H2	SWNT	--	Gas	Voltage	--	(171)
2007	Watts	O2, H2O,	MWNT	--	Gas	Resistance	--	(172)
2004	Valentini	O2, H2O, NH3, NO2	MWNT	--	Gas	Resistance	100 ppb	(173)
2011	Ghaddab	O3, NH3	SWNT	SNO2	Gas	Resistance	20 ppb, 1 ppm	(174)
2008	Zhao	Organic	SWNT	Pyrenecyclodextrin	Liquid	Resistance	--	(175)
2004	Picozzi	Ozone	SWNT	modeling paper		--		(176)
2010	Choi et al.	Pressure, Flow	MWNT	--	Gas	Resistance	--	(177)
2005	Suehiro	SF6	MWNT	--	Gas	Resistance/Partial Discharge	--	(178)
2010	Kang	SF6	SWNT	Benzene	Gas	Resistance	--	(179)
2011	Chaudhury	SO2, H2S	MWNT	PS	Gas	cyclic volt	--	(180)
2006	Lee	SOCl2, DMMP	SWNT	DNA	Gas	Resistance	50 ppm	(181)
2005	Auvray	TEA(aminocyclane)	SWNT	--	Gas	Voltage	20 ppb	(182)
2006	Wei	THF, ethanol, cyclohexane	MWNT	PVAc, PI	Gas	Resistance	--	(183)
2006	Parikh	Toluene, acetone, hexane, water	MWNT	PET	Gas	Resistance	--	(184)
2007	Consales	Toluene, xylene	SWNT	--	Gas	Optical, Acoustic	120 ppb	(185)
2001	Doleman	VOC	--	Polymers	Gas	--	1 ppm	(186)
2003	Hosseini	VOC	--	PVAc-graft-PPy	Gas	cyclic volt	200 ppm	(187)
2004	Penza	VOC	SWNT, MWNT	--	Gas	Acoustic	2 ppm	(188)
2005	Penza	VOC	SWNT	CdA	Gas	Acoustic	<1 ppm	(189)
2005	Snow	VOC, DMMP	SWNT	Pd	Gas	Capacitance	60 ppm	(32)
2006	Snow	VOC, DMMP	SWNT	--	--	Capacitance, Resistance	--	(190)
2005	Picaud	VOCs, other gases	SWNT	--	--	Resonant Cavity	ppb	(191)

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V EMERY  
2800 POWDER MILL RD  
ADELPHI MD 20783-1197

2 PROFESSOR ENRIQUE V. BARERRA  
(HCS) PADRAIG MOLONEY  
DEPT. OF MECHANICAL AND MATERIALS SCIENCE  
RICE UNIVERSITY  
PO BOX 1892  
HOUSTON TX 77251-1892